



**IN THE UNITED STATES PATENT & TRADEMARK OFFICE**

IN RE APPLICATION OF:  
SUGIURA ET AL.  
SERIAL NO.: 10/618,640  
FILED: JULY 15, 2003

:  
: EXAMINER: J. L. DOTE  
:  
: GROUP ART UNIT: 1756

FOR: EXTERNAL ADDITIVE FOR TONER FOR  
ELECTROPHOTOGRAPHY, TONER FOR ELECTROPHOTOGRAPHY,  
DOUBLE-COMPONENT DEVELOPER FOR ELECTROPHOTOGRAPHY,  
IMAGE-FORMING PROCESS USING THE TONER, AND  
IMAGE-FORMING APPARATUS USING THE TONER

**DECLARATION UNDER 37 CFR 1.132**

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Hideki Sugiura who deposes and states:

1. That I am a graduate of Shizuoka University, and received Master degree in Science in the year of 1992.
2. That I have been employed by Ricoh Company Limited for 14 years as a researcher of Analytical Chemistry, i.e., functional materials (1992-1999) and of Developer, i.e., a toner (1999 to the present).
3. That I am a co-inventor in the above-identified application.
4. That I have read and understood Barder et al. (US 4,983,369) and Inokuchi et al. (US 6,248,495), which have been cited against the claims in the above-identified application.
5. That oxide fine particles disclosed in Barder et al. and Inokuchi et al. do not satisfy a relation of:  $R/4 \leq \sigma \leq R$ , which is one of the requirements of the present invention.
6. That the following additional calculation and experiment were

conducted under my supervision on April 23, 2007, and during the period of from February 25, 2007 to March 21, 2007, respectively.

#### Calculation:

A standard deviation  $\sigma$  was calculated from the oxide fine particles shown in Fig. 2 of Barder et al. in the following manner.

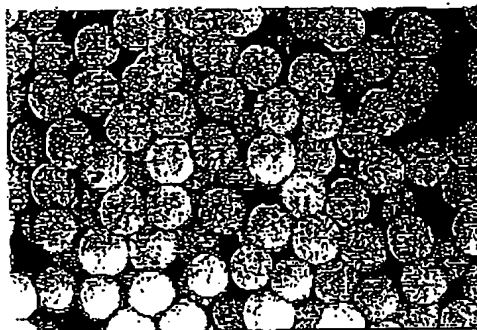
The image of Fig. 2 was scanned by a scanner (a product name: Imagio MP C4500, a manufacturer: Ricoh Company Limited), and was converted to a format of a bmp file. Thereafter, the images of oxide fine particles, i.e. silica particles, were selected by means of an image processing software (a product name: Photoshop 6.0, a manufacturer: Adobe Systems Incorporated). The selected images were analyzed by using Image-Pro Plus 4.5.1J manufactured by Media Cybernetics, Inc., and a perimeter of each oxide fine particle was obtained. Based on the obtained perimeters, a number average primary particle diameter of the oxide fine particles in Fig. 2 was calculated, and was found to be 1.13  $\mu\text{m}$ .

Sequentially, a standard deviation  $\sigma$  was calculated from the thus obtained average primary particle diameter by the following formula:

$$\sigma = \sqrt{\frac{1}{N} \sum_{i=1}^N (x_i - \bar{x})^2}$$

Note that, in the formula, N denotes a number of images of the oxide fine particles used for the calculation, i.e. 25 (see the figure below),  $x$  denotes a primary particle diameter of each oxide fine particle, and  $\bar{x}$  denotes the average primary particle diameter, i.e. 1.13  $\mu\text{m}$ .

FIGURE 1 Comparative Example, Two-Phase Reaction



As a result of the calculation, it was found that the standard deviation  $\sigma$  of the oxide fine particles shown in Fig. 2 was  $0.03 \mu\text{m}$ .

#### Conclusion of Calculation:

From the calculations above, it was found that the oxide fine particles shown in Fig. 2 of Barder et al. had a primary particle diameter of  $1.13 \mu\text{m}$  in number average, and a standard deviation  $\sigma$  of  $0.03 \mu\text{m}$ . Therefore, these oxide fine particles do not satisfy the relation of  $R/4 \leq \sigma \leq R$ , which is one of the essential requirements of the present invention.

#### Experiment:

Oxide fine particles were prepared in the same manner as described in Example 1 of Inokuchi et al. The details of the procedure are as mentioned below.

Methyltrimethoxysilane which had been purified by distillation was heated and nitrogen gas was bubbled therein. In this way, methyltrimethoxysilane was carried by nitrogen gas into an oxyhydrogen flame burner whereupon the silane was subject to combustion pyrolysis in the oxyhydrogen flame. At this point of time, the flow rate of methyltrimethoxysilane was  $1,268 \text{ g/hr}$ , the flow rate of oxygen gas was  $2.8 \text{ Nm}^3/\text{hr}$ , the flow rate of hydrogen gas was  $2.0 \text{ Nm}^3/\text{hr}$ , the flow rate of nitrogen gas was  $0.59 \text{ Nm}^3/\text{hr}$ , and the

spherical silica microparticulates received heat at a calorific value of 1.28 kcal/g. The spherical silica microparticulates formed were collected by a bag filter. A 5-liter planetary mixer was charged with 1 kg of the spherical silica microparticulates, and 10 g of pure water was added with stirring. After the mixer was closed, agitation was continued at 60°C. for 10 hours. The contents were cooled to room temperature, and 20 g of hexamethyldisilazane was added with stirring. After the mixer was closed, agitation was continued again for 24 hours. The residual reactants and ammonia formed were removed by heating at 120° C. and passing nitrogen gas. This yielded hydrophobic spherical silica microparticulates, i.e. oxide fine particles.

Thereafter, the thus obtained oxide fine particles were subjected to the examination under a scanning electron microscopy FE-SEM (a product name: S-4200, a manufacturer: Hitachi, Ltd.) at a magnification of 10,000 times. Using the thus obtained image file, images of oxide fine particles, i.e. silica particles, were selected by means of an image processing software (a product name: Photoshop 6.0, a manufacturer: Adobe Systems Incorporated). The selected images were analyzed by using Image-Pro Plus 4.5.1J manufactured by Media Cybernetics, Inc., and a perimeter of each oxide fine particle was obtained. Based on the obtained perimeters, a standard deviation  $\sigma$  was calculated in the same manner as described above. Based on the maximum perimeter, SF-1 and SF-2 were calculated by the following formulae:

$$SF-1 = (L^2 / A) \times (\pi / 4) \times 100$$

$$SF-2 = (P^2 / A) \times (1 / 4\pi) \times 100$$

Note that, in the formulae, L denotes an absolute maximum length of the oxide fine particle, A denotes a maximum projected area, and P denotes a maximum perimeter. If the particle is spherical, the values of both SF-1 and SF-2 become 100.

As a result, it was found that the oxide fine particles of Example 1 of Inokuchi et al. had a primary particle diameter of 80 nm in number

average, a standard deviation  $\sigma$  of 97 nm, SF-1 of 118, and SF-2 of 113.

**Conclusion of Experiment:**

From the results of Experiment above, it was found that the oxide fine particles of Example 1 of Inokuchi et al. had a primary particle diameter of 80 nm in number average, a standard deviation  $\sigma$  of 97 nm, SF-1 of 118, and SF-2 of 113. Therefore, these oxide fine particles do not satisfy the relation of:  $R/4 \leq \sigma \leq R$ , which is one of the essential requirements of the present invention.

7. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further deponent saith not.

Hideki Sugiura  
Hideki Sugiura

May 14, 2007  
Date